IRRADIATED GRAPHITE STUDIES PRIOR TO DECOMMISSIONING OF GI, G2 AND G3 REACTORS.

Jean-Pierre BONAL (1), Pierre-Philippe VISTOLI (2), Coryse COMBES (3)
(I)DENIDMNIS ALUI.M2E, CEA/Salay, 91191 Gif-sur-Yvette, France
(2)DENIDGESEEC, CEA/Cadarache, 13108 Saint-Paul lez Durance, France
(3)DENIDECISA30 RC, CEAICadarache, 13108 Saint-Paul lez Durance, France

GI (46 MW*), G2 (250 MW*) and G3 (250 MW h) are the first French plutonium production reactors owned by CEA (Commissariat à l'Energie Atomique). They started to lie operated in 1956 (GI), 1959 (G2) and 1960 (G3); their final shutdown occurred in 1968, 1980 and 1984 respectively. Each reactor used about 1200 tons of graphite as moderator, moreover in G2 and G3, a 95 tons graphite wall is used to shield the rear side concrete from neutron irradiation. GI is an air cooled reactor operated at a graphite temperature ranging from 30°C to 230°C; G2 and G3 are C02 cooled reactors and during operation the graphite temperature is higher (140°C to 400°C). These reactors are now decommissioned, but the graphite stacks are still inside the reactors. The graphite core radioactivity bas decreased so that a full decommissioning stage may lie considered. Conceming this decommissioning, the studies reported ere are: (i) stored energy in graphite, (ii) graphite radioactivity measurements, (iii) leaching of radionuclide (14C, "CI, Ni, 60Co, H) from graphite, (iv) chlorine diffusion tbrough graphite.

Stored energy offert referred as Wigner energy, occurs in neutron irradiated graphite because carbon atoms are displaced from their normal lattice positions into configurations of higher potential energy. During irradiation, simultaneous thermal and irradiation annealing takes place, but there is a net energy gain which depends on both eutron fluence and irradiation temperature. For irradiation temperatures above 350°C the accumulation of stored energy is negligible. Below 350°C significant amount of Wigner energy can lie accumulated, but at irradiation temperatures below 150°C, the amount of energy is larger and may lie released at low temperature. If the graphite is heated to a tbreshold temperature (0.), about 30°C above its irradiation temperature, the stored energy begins to release, a temperature of 1800°C is required to release all the Wigner energy. An irradiated graphite will lie thermally stable if, at any temperature the differential enthalpy (dH/d8) is lower than the specific heat (C,) of unirradiated graphite; in this case no self heating can occur. In order to decrease the amount of stored energy, the graphite stack of GI reactor underwent 18 annealing operations during its life; the last annealing occurred 2.5 montbs before its final own. Alter the final shutdown, a few stored energy measurements were carried out. For graphite irradiated at the er fluence (7 1020 n.cm 2 cp FG) and at 76°C, the energy released between 20°C and 800°C (II20-800^c) is 723 J.g', however at any temperature (dHIdO) < (C,), so no self heating can occur. Alter the G2 final shutdown, stored ener, bas been measured on several samples trepanned at différent places in 8 charnels. The higher value of H20-ew=c is 635 J.g-'; it bas been measured for a sample irradiated at 250°C/3.2 1021 n.cm 2 cp FG. However, for all the samples and a temperature (dHId8) is below (Cp). No Wigner energy measurement of G3 graphite bas been carried out alter its finit but results obtained 9 months before (September 1983), showed a similar behaviour to that of G2 graphite.

raphite stack, the total activity of y-emitters in 20 GBq, about 80% of this activity is due to 60 Co d 154Eu. The total activity of '4C i estimated to 7750 GBq. There no activity results for others (3" emitters (-60, 63Ni, 'H) for GI graphite. Three drillings (axial, vertical and horizontal) in the G2 graphite stack have been carried out alter its final shutdown. In the moderator zone, the average y activities in 2005 of the samples trepanned in the axial, vertical and horizontal drillings are 2.8 kBq.g' (95% 60Co), 2 kBq.g-' (87% 'Co) and 1.2 kBq.g-' (78% 'Co) respectively. As regards to (3- emitters in the G2 moderator zone, their average activities in 2005 are 163 kBq.g-' (89%3 H and 9% 14C) in the horizontal drilling and 90 kBq.g' (57% 3H and 43% 14C) in the vertical drilling. The 14C activity distribution fits closely with the neutron flux distribution, unlikely the tritium distribution is uniform in the graphite stack. The average a activities in 2005 are about 10 Bq.g-' in the moderator zone and 2 Bq.g-' in the reflector zone. No drilling occurred in G3 graphite stack alter its final shutdown, therefore there is no radioactivity data.

Graphite leaching data are interesting either for graphite sub-surface repository site studies or in case of graphite core dismantling under water. Leaching experiments on a graphite core extracted from a G2 reactor brick show that chlorine moves outside very quickly and is not retained in the matrix while 14C, mostly produced by 14N, is likely located within the closed porosity structure or bound to the surface atoms in the regions accessible to nitrogen. Leaching results were obtained with 100 g; 18mm thick samples dip into différent leaching solutions. They show that the main part of the chlorine content is released in less then three montbs (90% in about 15 days). Tt means that the main part of chlorine is easily reached by leaching water and also that it is not well linked inside the carbon matrix. On the contrary, 14C is very little leached, actually, the leachate analysis are often below the detection limit. The value of leached 14C related to initial content can lie estimated to about 10' a year. The leachability of nuclides, less important terni behaviour such as 63Ni, 60Co, 3H, bas also been determined.

Diffusion experiments involving chlorine through inactive slices of G2 graphite have been performed. The results show that the diffusivity constant can reach up to 5 10"12 m2.s' which is ahnost as high as the one obtained for tritiated water. This hi¢h diffusivity is in eood accordance with the results of leachin¢ reported above.